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AN APPARATUS FOR THE PRODUCTION OF LARGE METALLIC CRYSTALS BY SOLIDIFICATION AT HIGH TEMPERATURES

by

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INTRODUCTION

The commercial production of large single crystals is rapidly making available specimens of inorganic compounds for research purposes and special applications. The Harshaw Chemical Company is growing halide crystals in pound denomination[1]; the earlier researches of Stockbarger are being used to good advantage.[2] The Verneuil Method for preparing synthetic sapphires[3] has been developed by the Linde Air Products[4] who are producing not only large sapphires, but a number of other refractory oxides. Considerable interest in the problem of growing adequate sized piezoelectric quartz crystals has been manifested, and progress is being made in this direction.[5]

The literature contains ample evidence of extensive investigations pertaining to the preparation and properties of metallic crystals. In general, only the lower melting metals such as zinc, lead, cadmium, etc., have been grown with some consistency with dimensions that might be considered as appreciable[6]; copper and certain alloys (gold-copper, alpha and beta brass, for example) have been prepared in sizes of some note,[7] but beyond the range of approximately 1100°C, the results are quite sporadic.[8]

Although large crystals of metals having high melting points can be prepared by other methods* which avoid high temperature complications, solidification from the melt apparently is most amenable to large scale operation.[10]

Recently there has developed as a result of war-time researches marked awareness of the need for large metallic crystals.[11] In particular the desirability of suitable delay lines and similar storage devices[12] have intensified the importance of such materials.[13] This paper is an outgrowth of these developments.

The apparatus to be described evolved from the desire to fulfill the considerations of having a relatively simple arrangement for growing large crystals of metals with design features which would allow for reasonable flexibility. The various schemes for growing crystals from the gaseous, liquid, solid, and solution states were critically re-examined and the decision to prepare the specimens from the melt made.

THE APPARATUS DESIGN AND CONSTRUCTION

The sundry variations of growing the crystals from a melt are all based on the underlying process of controlled solidification with due observance of the principles of nucleation and growth.

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*The other methods referred to are the strain-anneal and electroplating techniques. Metals having allotropic transformations between room temperature and their melting points may be thus most effectively grown as large crystals.

The techniques of Bridgman,[14] Stoeber,[15] Czochralski,[16] and Obreimov-Schubnikov[17] were carefully evaluated, and the Bridgman Method finally selected because of its apparent popularity. Descriptions of apparatus utilizing the Bridgman Method, some of which are rather crude and others which are quite elegant in their construction, may be found in the literature.[18] The vacuum furnaces used by Nix[19] and Siegel[20] were considered as best representative of what could most probably serve effectively for most metals since the exclusion of air is generally mandatory. The wire suspension arrangement for raising and lowering the specimens into and out of the furnace was thought to be inadequate for large melts of reactive metals; the method of moving the melt on a mercury column which had been used by both Quimby, et al.,[21] and Gwathemly and Benton[22] in different forms was further modified as will be described.

Figure 1 shows the equipment which has been used to prepare a wide variety of large metallic crystals; copper specimens of the order of 2 inches in diameter and 6 inches in length, for example, have been produced with relatively little difficulty. Metals and alloys in the range from 1100°C to 1600°C have also been grown in reasonable denominations with the basic limitation of resistance type furnaces restricting operation beyond this range. Although the apparatus has been limited to vacuum operation, it can readily be adapted to usage with inert atmospheres if desired as would be the case when dealing with highly volatile metals. The apparatus will not be described in terms of its basic components, which are: (a) the vacuum system, (b) the furnace chamber, (c) the furnace unit, (d) power supply and temperature control, and (e) the lowering-raising mechanism.

The Vacuum System

Gasket seals, waxed joints, and stop-cocks were eliminated wherever possible, particularly on the high vacuum side to minimize leak troubles; most of the joints were hard soldered or welded. A vacuum of 5×10^{-6} mm could be maintained with the usual arrangement of Welch Duo-Seal two-stage fore-pump and Distillation Products three-stage vertical diffusion pump. In the course of a run, however, severe out-gassing would reduce the vacuum to approximately 10^{-3} mm. Because of the wide variations in gas composition, it was found that the standard gauges were not serviceable in this range. Instead, a Lepel High Frequency Oscillator (commonly known as a spark-coil) was used to gain an approximate estimate of the pressure; in any event, it could detect if and when a leak occurred.

Furnace Chamber

Figure 2 shows the furnace chamber extending to the bottom of a mercury well (33) on one end and sealed at the top by a steel cover and gasket (7), (8) The steel sidearm pumping lead, hard soldered to the chamber, is bolted to the diffusion pump and the seal made via an indium metal gasket.* There are several openings on the chamber. Three windows (21) each at a different level provide for sighting. These were not used in any of the runs made, but should prove of value in studying solidification or other processes at much lower temperatures. These windows are made by a special process and are adequately described in the Tube Shop Handbook of the M.I.T. Radiation Laboratory. Their novel feature is the distortionless view one sees through them, which is of material importance in many instances. Below the bottom window is another opening (22) through which a thermocouple (23) can be inserted. The outside of the chamber is wrapped with approximately 150 feet of 3/8-inch copper tubing (9) which is soft soldered into place; the water cooling provided will dissipate more than 5kw with the chamber wall running a bit warm. A steel ring (25) was welded into place on the inside to support the furnace unit.

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*Indium is better than lead for this purpose because it is softer and less inclined to work-harden.

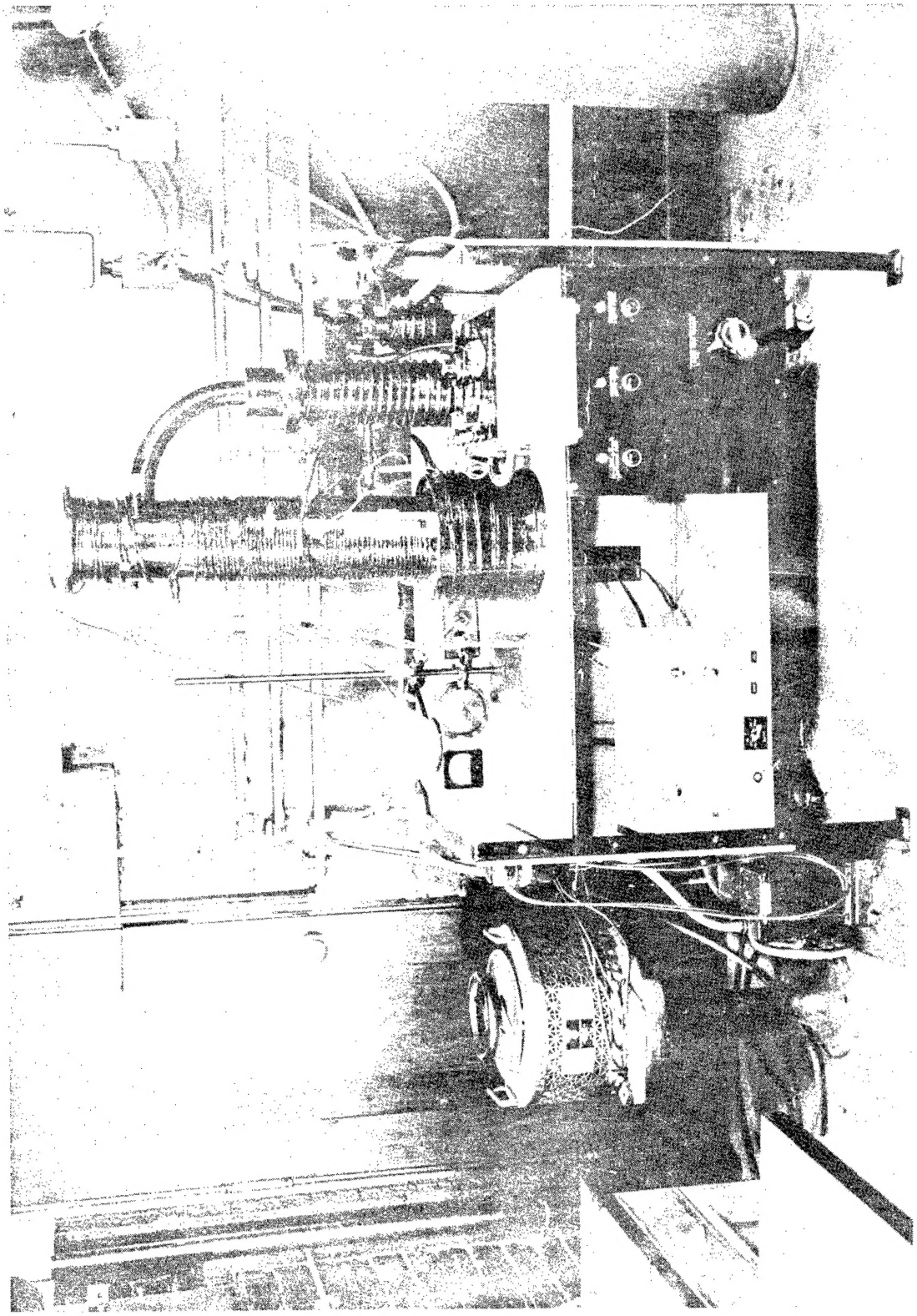

Figure 1. General view of apparatus.

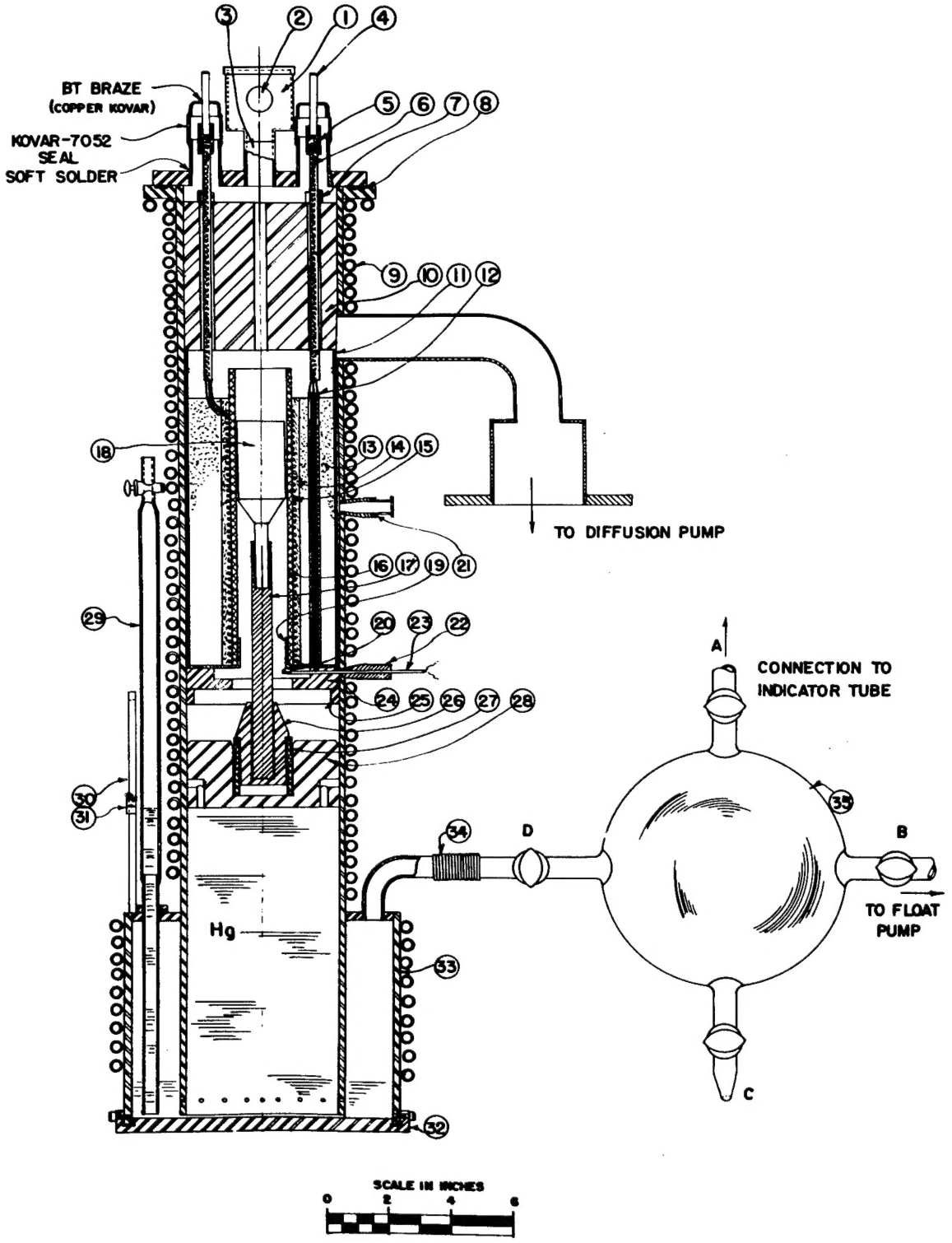

Figure 2. Sectional view of furnace chamber.

Furnace Unit

Because of the high temperatures and prolonged periods of operation, and because of the gradient requirements necessary for single crystal propagation, the furnace design proved to be most formidable; the difficulties appeared to be most marked when attempting to grow the larger sized specimens. Also there were complications resulting from vapors attacking ceramics and the general misbehavior of the insulation at high temperatures.

An alundum core (16) RA-98 or RA-1139 (supplied by the Norton Refractory Company) is wound with the required number of turns of molybdenum wire (15) of predetermined diameter. A layer of special grade alundum cement (silica free) (14) is applied over the windings and pre-fired up to 300°C. The current carrying capacity of the return leads is enhanced by using three strands of wire, each of which is encased in an individual alundum tube (12) for protection. The core is slipped over the lip (19) of a steel bucket (11) which is then filled with coarse-grain, silica-free, alundum sand (13). A pair of holes close to the upper rim of the bucket allows for the ready insertion and removal of this unit by a pair of steel hooks. The bare wires protruding above the level of the sand are inserted into a stainless steel sleeve (6) and held firmly in place by a number of set-screws; this arrangement provides the necessary rigidity for facilitating the job of making electrical contact in a vacuum and also serves to protect the embrittled "moly" leads from repeated handling.

The stainless steel sleeves pass through a lavite plug (10) which centers them and also offers insulation above the furnace. Two lock nuts (7) fasten the power-leads in place. A pair of connectors (5) are fastened to the power-leads by set-screws. Between steel bucket and ledge is a section of lavite insulator (24) which is braced by a thin steel section not shown.

Power Supply and Temperature Control

Evidently, temperature control in high power, high temperature furnaces cannot be handled in conventional fashion.[23] For temperatures over 1000°C thermocouples cannot be relied upon for dependable performance. There are two recommended approaches:

(1) Control thermocouple at some convenient point in furnace below 1000°C, but so arranged as to give temperature control in the hot zone which may be well above this temperature.

(2) Maintenance of constant power by using a voltage-stabilized supply.

Because of the uncertainty of the nature of the gradients within the furnace, method 2 seemed preferable; for a furnace relatively free of gradients method 1 is inherently capable of more accurate control, but method 2 will generally be adequate since in the range of temperatures of present interest, temperature control better than a few degrees is practically meaningless, primarily because of the limitations of the temperature measuring devices.[24]

A General Radio 7 KVA variac rated at 220 v and 30 amp was operated off a two phase 60 cycle 220 v 30 amp line. The power was introduced into the vacuum furnace by means of the power leads (4) indicated in Figure 2. These leads were made up as a unit which was then soft soldered into the steel cover plates. They are very sturdy in nature and can be made to sizes which will carry currents of the order of hundreds of amperes. A long section of 705-2 glass permits one to see the contact made. For high vacuum operation the contact is best made by dry fits, but for low vacuum or inert atmosphere operation mercury contacts may be used.

The temperatures were read with a Leeds and Northrup Optical Pyrometer through the 1-inch window (3) by sighting a glass prism (2) in the brass housing (1). In actual practice, no apparent temperature fluctuations were observed during the course of operation of the furnaces. The thermal inertia of the furnace was such as to eliminate rapid fluctuations of temperature even though the voltage were to fluctuate for any reason. Moreover, some approximate data taken on the voltage-temperature characteristics of the furnace indicated that a one volt change would result in a

temperature change of only a few degrees. Still another indication of the temperature regulation possible was shown by the ability to hold a constant temperature on some particular object in the furnace for hours at a stretch by merely setting the voltage at a given level.

The Lowering-Raising Mechanism

Reference to Figure 2 will indicate the arrangement used to vary the position of specimens in the furnace chamber. The principle of altering the height of a mercury column which was used successfully by Quimby and independently by Gwathemly and Benton was carried out by the former by draining off mercury through a rotating stop-cock into an evacuated chamber and by the latter simply by varying the height of a leveling bulb which was slowly lowered by a gear reduction system.

The present design is thought to have advantages over the aforementioned techniques; the height of the mercury column is varied by regulating the pressure in the steel reservoir (33). This was done by a sequence of manipulations on the stop-cocks attached to the volume tank (35); bellows (34) were inserted as indicated to minimize the stresses on the glass connections during the various stages of manipulation. An indicator tube (29) provided with a scale and lens viewer (30) and (31) was used to determine the height of the column (and thus the position of the specimen).

A uniform rate of lowering is possible only under certain conditions, foremost of which is the maintenance of relatively lower pressures in the volume tank than in the reservoir. The uniformity of lowering was checked up to rates of approximately 6 mm per minute, at which point slight fluctuations of the kind shown in Figure 3 were noted. Since the rates used in the crystal growing runs were never greater than 2.5 mm per minute, the lowering rate was uniform; exceedingly slow rates of the order of mm per hour were not amenable to careful regulation because of stopcock difficulty —it was found important that stopcock D be greased properly if good performance were to be had. Grease or dirt in the stop-cock hole will give trouble. This possible source of interference could be readily eliminated by inserting a T-branch between the reservoir and the volume tank; one branch could be used for evacuation and raising purposes, while the other could have a metal capillary valve for the lowering process. This suggestion is made for future experimentation.

The steel float (28) is inserted into the chamber prior to the installation of the mercury and the bottom plate (32). The float design was carefully considered in terms of the amount of tilting which would be experienced by melts supported at some distance from the face of the float. If the fit between the float and chamber is too close, there is a tendency for sticking and general erratic behavior; on the other hand, if too loose, the tilting effect is enhanced. Thus, it was necessary to make a simple analysis of the variation of the amount of tilting with the degree of clearance between the float and the furnace chamber. A working formula which applies for the conditions prevalent is

$$\delta = \frac{L(W - w)}{h}$$

where delta (δ) is the amount of horizontal displacement,

W is the inside diameter of the furnace chamber,

w is the diameter of the float,

L is the length of the crucible support plus crucible length, and

h is the height of the float.

For the setup used, calculation based on the formula indicated that a clearance of several thousandths should not give significant values for delta (δ). It is also a safe precaution to have the inside wall of the chamber free of scratches and bumps.

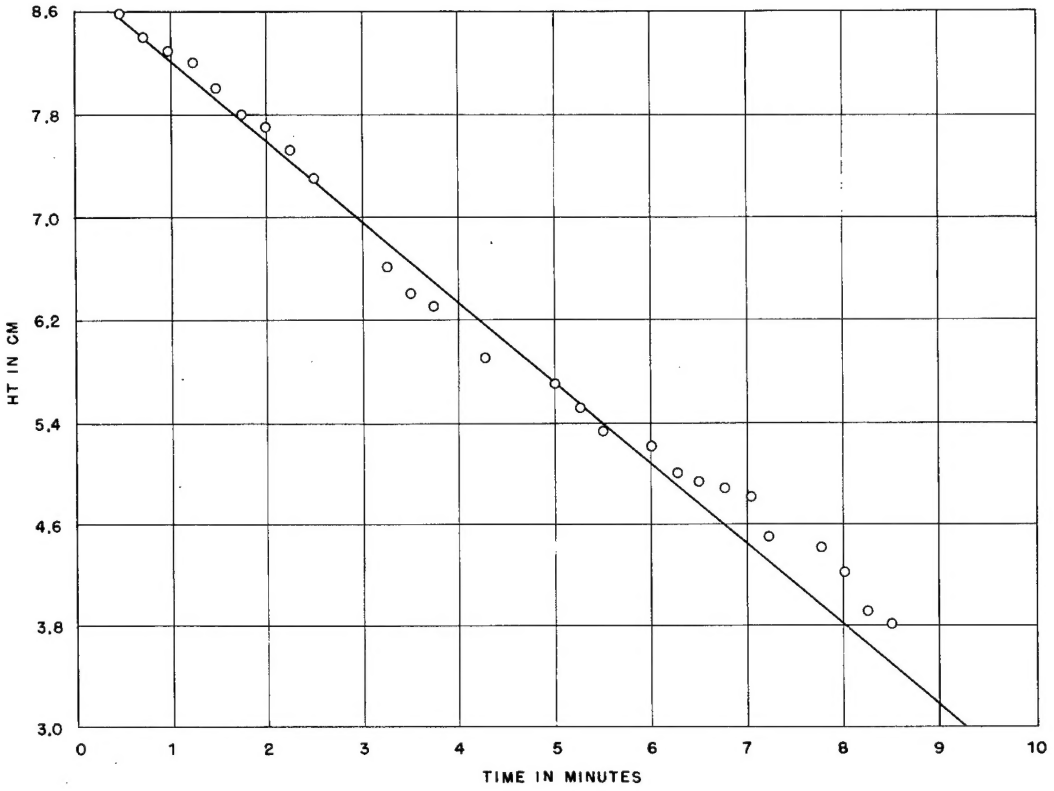

Figure 3. Typical plot of mercury level vs time showing fluctuations at rapid lowering rates.

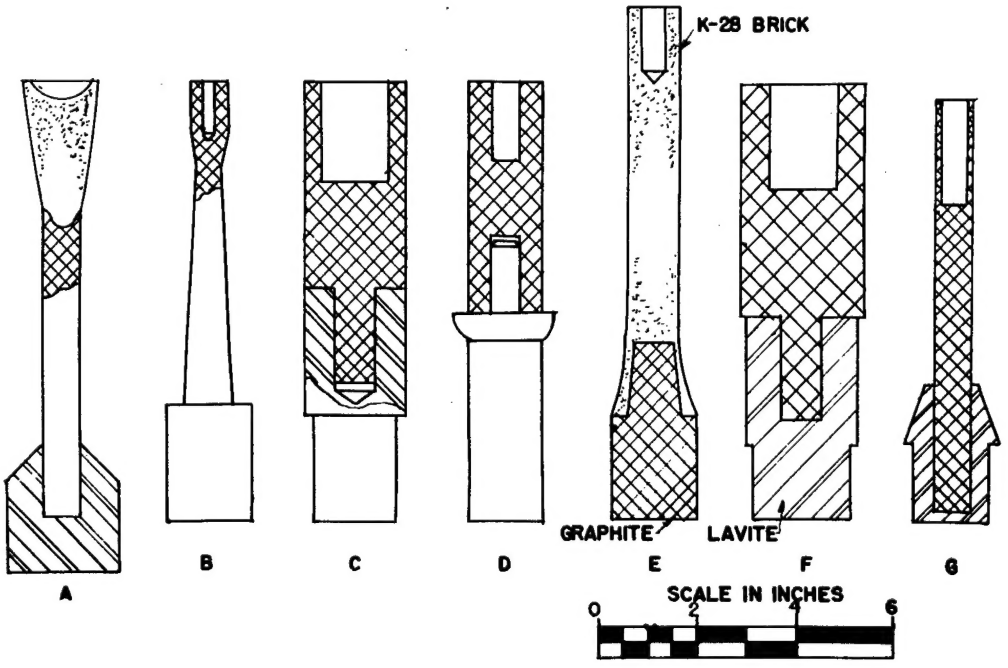

Figure 4. Pedestal design.

Figure 2 shows a particular pedestal arrangement (17), (20), and(27). In the course of developing growth techniques for various metals and alloys a variety of pedestal designs were devised. These are shown in Figure 4. The shapes and materials of construction were altered to modify the temperature gradients in the charge. In general, they were composites of graphite, firebrick, and lavite.

To achieve the proper levels of the specimen, both during the melting operation and the final solidification state, it was necessary to select the proper relative dimensions for the reservoir and the furnace chamber. Some degree of flexibility was required to accommodate the different sizes and weights of the many specimens; this was accomplished by varying the amount of mercury in the reservoir. The mercury can be siphoned in and out through the indicator tube as the situation might demand. Occasionally the indicator tube, as well as the mercury in it, would become sufficiently dirty to make readings difficult by virtue of the meniscus becoming anomolous and the general obscuring of its position. The tube and mercury were readily cleaned from time to time by admitting dilute nitric acid, water, and acetone in proper sequence via the stopcock shown. It is important to eliminate all traces of water or acetone in the tube prior to starting up again for operation; if this is not done, bubbles will form beneath the mercury in the tube to interfere with proper operation by rising to the surface when a run is in progress.

OPERATIONAL PROCEDURE

Having described the individual design aspects of the components, we will now delineate the manner in which the apparatus may be used for a typical run. By no means are the following details unique—a broad range of variations is possible and, as a matter of fact, additional modes of operation have been utilized. The operational sequences can conveniently be classified as: (a) loading, (b) starting-up, (c) outgassing and pre-heating the furnace, (d) raising and melting the specimen, (e) lowering the specimen, and (f) finishing the run.

Loading

The specific nature of the run determines the kind of pedestal, crucible, and furnace to be used. The pedestal is inserted into the hole of the steel float with the crucible or crucible holder in place; the maximum and minimum position of the crucible is then checked by raising and lowering the mercury column. The column is raised by connecting stopcock A or C to a compressed air line and then keeping the proper cocks closed or open. Lowering is accomplished by simply letting the compressed air in the volume tank escape. The maximum and minimum position of the crucible in the furnace can be adjusted by varying the quantity of mercury in the reservoir and the height of the pedestal. Different sizes and denominations of specimens require that these adjustments be made.

With these preliminaries dispensed with, the furnace is then lowered onto the lavite insulator above the steel shelf by means of a pair of steel hooks. The task of getting the crucible into the furnace opening is facilitated by having the float completely lowered before the furnace is inserted. Having set the furnace into position, one can again check the maximum and minimum positions of the crucible.

The lavite plug is then slipped over the protruding stainless steel connecting rods and lowered into position by a pair of pointed tongs inserted into the sighting hold. The locknuts are put into place to hold the connector-rods rigidly in place and the connector-cups fastened into place.

The gasket on top of the furnace chamber is well greased and the steel cover carefully lowered into place as the power-leads make contact with the connector-cups. The steel cover is subjected to a slight rotary motion which serves to assure good contact and to spread the grease film under it to effect a good seal.

Starting Up

A rubber hose connection is made between the indicator tube and stopcock A on the volume tank. The float pump is then started up and the mercury column slowly raised up the entire length of the indicator tube and just beyond the opening of the stopcock to eliminate all the air. The indicator tube is closed off and the hose connection removed. The volume tank is next evacuated with stopcock D on the closed position.

At this point the fore-pump is turned on and the furnace chamber evacuated. To prevent the mercury column rising too far into the chamber, it is important to balance the pressures on both sides of it. This is accomplished by noting when the mercury in the indicator tube begins to fall and as it does, stopcock D is slowly opened to decelerate the downward motion of the mercury column. With both sides of the mercury column evacuated, the diffusion pump is then turned on, followed by the water cooling system.

Outgassing and Pre-Heating the Furnace

The power supply is connected to the furnace leads, and the power gradually increased. Too rapid heating is avoided in order to prevent thermal stresses from causing cracking of the furnace core or crucible. A furnace being used for the first time gives off voluminous amounts of water vapor and carbon dioxide. A new furnace is generally outgassed overnight at a dull red heat. Subsequent usage of the furnace will not require outgassing periods of long duration. The furnace is heated above the maximum point of operation to be effective in the melting operation to drive off gas that might otherwise be slowly evolved at the lower operating point.

Raising and Melting the Specimen

The specimen is slowly raised into the furnace by careful manipulation of the stopcocks on the volume tank. The raising process is watched through the window on the steel cover via the right angle prism in the brass case. When the specimen has been raised to desired height (as shown by the position of the mercury level in the indicator tube), stopcock D is closed and the volume tank evacuated in readiness for the lowering operation.

Temperature readings are taken periodically to follow the progress of the melting operation. If the power required to effect melting has not been predetermined in some preceding run, the voltage is gradually increased until melting is noted. Since some specimens will not show a clear liquid melt,* it is of considerable help to plot the temperature readings versus time and get a heating curve from which the melting halt can be detected. After some experience, the halts can be noted without the necessity of plotting the data, but in some instances this cannot be dispensed with.

The temperature readings are subject to a number of sources of error and must therefore be corrected.[26] Where a clear liquid surface can be observed (as in the case of copper, for example) it is not unusual to experience an apparent drop in temperature although actually the temperature has increased. In some instances the drop has been as much as approximately 150°C. This phenomenon can be attributed to the marked change in the spectral reflectivity at the melting point.

To minimize possible reactions of the melt with the crucible or even of the crucible with the holder, and also to minimize evaporation, it is important to avoid overheating the molten change. Yet, one must be certain to have effected complete melting.

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* Reactive metals such as aluminum, beryllium, etc., form oxide skins even in a high vacuum. The oxide skin on the surface of the melt obscures the nature of the charge below.

Lowering the Specimen

Having melted the specimen and allowed sufficient time for the elimination of any solidus present in the melt, the solidification process is initiated. The temperature is stabilized at approximately 100°C above the melting temperature. Lowering the specimen out of the furnace is then begun by cautiously turning stopcock D to the open position; the volume tank must first be completely evacuated to the point where no glow discharge can be excited with a spark coil. Air escaping into the volume tank as stopcock D is opened will give rise to the glow discharge and thus indicate that lowering has commenced. The opening of the stopcock is adjusted until the desired rate of lowering is achieved. This is done by a series of alternating back and forth maneuvering, sufficient time being allowed for equilibrium to set in for each stopcock setting.

Finishing the Run

When the specimen has reached the minimum position, or when solidification is known to be complete, the power is gradually reduced. Reducing the power too rapidly may cause cracking of the furnace core or other ceramic materials in the hot zone. With the power completely off it may take hours for the furnace to cool, particularly if it is well insulated. So to accelerate the cooling process the specimen may be raised back into the furnace after the furnace has cooled below the melting point region. This step will simultaneously anneal the specimen and remove thermal stresses that might have been left by the original gradient condition that existed during the solidification.

The cooling operation completed, the usual operation of shutting the diffusion pump and the fore-pump is followed by admitting air through the bleeder-valve on the steel cover. It has been found advisable not to attempt to remove the specimen while the furnace is still in place. By removing the furnace first, one can carefully extract the crucible containing the specimen.

DISCUSSION

In the course of the numerous runs made with the apparatus, the complexities arising from prolonged, large scale operation at high temperatures were made evident. A number of these are directly attributable to the presence of a metallic charge and others originate because of the inherent difficulties of high temperature operation.

The crucible problem for metallic melts is much more severe than for inorganic compounds. The technique of thin-walled metal crucibles devised by Stockbarger[2] cannot be utilized because of obvious alloying effects. Refractory crucibles made of alundum, beryllia, graphite, etc., when in contact with molten metals for prolonged periods will react or show marked penetration. This gives rise to contamination and sticking; silicon, for example, in an alundum crucible will not come out readily and the crucible needs to be broken away. The need for high gradients also causes difficulty with crucibles by favoring thermal stress cracking.

Resistance-wound furnaces are a potential source of trouble because of a number of deleterious effects, foremost of which is electrical breakdown at elevated temperatures. This can be reduced somewhat by designing the furnace for low-voltage, high-current operation; one cannot carry this process too far because of the wire size limitations. Resistance-wound furnaces are also subject to attack by metal vapors. Some degree of minimization of this difficulty is possible by proper shielding of the furnace walls. One cannot, however, completely obscure the surface of the charge because of the necessity of sighting it during the course of a run.

The longevity of such furnaces is seriously handicapped by gradual evaporation of the resistor element and increasing embrittlement of the furnace core. The presence of high gradients is certainly not beneficial.

There appears to be some evidence which indicates that the Bridgman Method may not be the best means for introducing the gradient into the molten charge. The large cross-sectional area of melts with relatively high thermal conductivity tends to level off the gradients existing at the bottom of the furnace. For this reason it was found necessary to support the crucibles on graphite pedestals in order to create sufficiently high thermal gradients in the charge. The graphite pedestals, however, markedly increased the power requirements. It was found necessary to use combinations of graphite and insulators such as lavite and firebrick to compromise between power-loss and thermal gradients. The Obreimon-Schubnikov technique of instituting the gradient after melting has taken place by introducing a heat sink was surmised to be a more efficient procedure. Some preliminary attempts in this direction were quite encouraging.

The relatively higher degree of impurities in the metal specimens further distinguishes the growth of metallic crystals from inorganic crystals. Insoluble matter in the metal will settle out when the specimen is kept molten for some time. The presence of the insoluble impurities is probably responsible for the observance of an optimum rate of solidification for producing large crystals.

It was generally evidenced that increasing the dimensions of the specimens not only altered the optimum conditions, but also augmented the tendency of parasitic grains appearing. Although a number of specimens were not truly single crystal, i.e., such parasitic grains were evident, their general usefulness was not seriously interfered with. Thus, the term single crystal was avoided in the title of this paper for the sake of completeness and accuracy; moreover, the distinction between the true single crystal and the mosaic crystal is not well enough defined to permit one to use the terminology of single crystal with impunity.

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REFERENCES

1. Kremers, H. C., Chemical and Engineering News, July 7, 1947.
2. Stockbarger, D. C., Rev. Sci. Instr. 7:133-136 (1936); Jour. Optical Soc. 27:416-419 (1937).
3. Verneuil, M. A., Ann. de Chemie et Physique 8:320-48 (1904).
4. Linde brochure (1946) "Synthetic Sapphire, Ruby, and Spinel."
5. Hale, D. R., Science 107:393-394 (1948).
6. Cinnamon, C. A., Rev. Sci. Instr. 5:187-190 (1934). Hasler, M. F. ibid., 4:656-660 (1933). Betty, B. B., Amer. Soc. Testing Materials, Proc. 35:193-200 (1935) II. Miller, R. F., Trans. A.I.M.E. 111:135 (1934).
7. Hausser, K. W., and P. Scholz, Wiss. Ver. Siemens-Konzern 5:144-164 (1927). Siegel, S., Phys. Rev. 57:537-545 (1940). Rinehart, J. S., Phys. Rev. 58:365 (1940). Webb, W., Phys. Rev. 55:297-305 (1939).
8. Van Liempt, J. A. M.,Proc. A.I.M.M.E. Tech. Publ. 15:358 (1927). Kaya, S., Sci. Rep. Tohoku Univ. 17:1157-1177 (1928); ibid., 17:839-883 (1928).

9. Carpenter, H. C. H., Metal Industries 44:557-560, 584-587, 637-638 (1934). Erdly-Gruz, T., and R. F. Kardos, Zeits. f. Phys. Chem. 178:256-265 (1939).

10. Kurylenko, C., Rev. Opt. (Theor. Instrum.) 23:1-19 (1944).

11. Arenberg, D. L., M.I.T. Rad. Lab. Report 932 (1946). Also see J. Acous. Soc. Am. Vol. 20: 1-26 (1948).

12. TRE Report T-1539, August 1943, "Preliminary Experimental Enquiry into the Possibility of using Solid Materials for Supersonic Transmission Media for Delay Cells and other Devices."

13. Emslie, A. G., and R. L. McConnell, "Radar System Engineering," Vol i, Chap. 16, McGraw-Hill Book Company, Inc., New York,1947.

14. Bridgman, P. W., Proc. Amer. Acad. Arts and Sci. 60:305-383 (1925); ibid, 63:351-355 (1929).

15. Stoeber, F. Zeits. f. Krick. 61:299-314 (1925).

16. Czochralski, J., Z. Physik. Chem. 92:210-221 (1917). J. Chem. Soc. 112: II 302 (1917).

17. Obreimov, I. and L. Schubnikov, Zeits. Physik 25:31-36 (1924).

18. Hoyem, A. G., Proc. Iowa Acad. Sci. 36:301 (1929).

19. Nix, F. C., Rev. Sci. Instr. 9:426-427 (1938).

20. Siegel, S., Phys. Rev. 57:537-545 (1940).

21. Quimby, S. L., Phys. Rev. 39:345-353 (1932).

22. Gwathemly, A. T., and A. F. Benton, Jour. Phys. Chem. 44:34-42 (1940).

23. Stansel, N. R., Industrial Electric Heating, Wiley and Sons, Inc., 1933.

24. American Institute of Physics, "Temperature, Its Measurement and Control in Science and Industry," Reinhold Publishing Corporation, New York, 1941.

25. Marks, L. S., Mechanical Engineers Handbook, "Flow of Compressible Fluids," pp. 353-361, McGraw-Hill Book Company, Inc., New York 1941.

26. Forsythe, W. E., Measurement of Radiant Energy, McGraw-Hill Book Company, Inc., New York, 1941.

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